ELSEVIER

Journal of Crystal Growth 241 (2002) 231–234

JOURNAL OF CRYSTAL GROWTH

www.elsevier.com/locate/jcrysgro

Analytical solution to Matthews' and Blakeslee's critical dislocation formation thickness of epitaxially grown thin films

A. Braun[a,*], K.M. Briggs[b], P. Böni[c]

[a] *Ernest Orlando Lawrence Berkeley National Laboratory, Environmental Energy Technologies, One Cyclotron Road, Berkeley, CA 94720, USA*

[b] *BTexact Technologies, Martlesham Heath, IP5 3RE Suffolk, UK*

[c] *Technische Universität München, Physics Department E21, James-Franck-Straße, D - 85748 Garching, Germany*

Received 1 October 2001; accepted 7 January 2002
Communicated by J.J. Derby

Abstract

For the first time, an analytical expression for the critical thickness for the onset of misfit dislocations as established by Matthews and Blakeslee is presented. It is the so-called *Lambert W* function which reflects the curvature of this critical thickness. With the arrive of the analytical solution, expressions of arbitrary complexity that involve the critical thickness can be handled much more easily. Its practical application is demonstrated by implementation of Vegard's rule. © 2002 Published by Elsevier Science B.V.

PACS: 02.30.Ik; 68.55.−a; 81.15.−z; 81.70.−q

Keywords: A1. Computer simulation; A1. Critical thickness; A1. Defects; A2. Growth models; A3. Atomic layer epitaxy

1. Introduction

About 25 years ago, Matthews and Blakeslee [1] introduced a model for the onset of misfit dislocations in thin films, when epitaxially grown on single crystal substrates. This model is based on thermodynamic assumptions and represents an elasticity theory approach.

Despite the fact that experimental data often deviate from predictions made by the model, the majority of researchers appreciates this model, probably due to its simplicity and conceptual transparency. Since then, their article has been cited almost 2000 times [2], which gives a clear indication about the importance of their pioneering work. However, the model was deficient insofar as it did not provide an explicit expression for the critical film thickness d_c, but an implicit one of the type:

$$d_c = A \ln(Bd_c), \tag{1}$$

*Corresponding author. University of Kentucky, Consortium for Fossil Fuel Science, 111 Whalen Building, 533 South Limestone Street, Lexington, KY 40506, USA. Fax: +1-859-257-7215.

E-mail address: abraun@engr.uky.edu (A. Braun).

0022-0248/02/$ - see front matter
PII: S0022-0248(02)00941-7

where A and B represent material constants. Although Eq. (1) is very easy to solve by an iterative numerical method, this is not necessary; the exact solution of Matthews' and Blakeslee's equation is given by the so-called *Lambert W* function [3].

The transcendental W-function was postulated by the 18th century scientist Lambert [3], a colleague of mathematician Leonhard Euler, to solve the equation

$$W(k)\exp W(k) = k. \tag{2}$$

The W-function allows for the explicit solution of entire classes of differential equations, which to date only could be solved numerically, and is experiencing today a renaissance in various fields of science and engineering [3,4], not least in the modeling of thin film growth [5,6].

2. Results

In many cases, when a thin film is epitaxially grown on a planar substrate, the film material adopts the crystal lattice of the substrate. When the bulk structures of the film material and the substrate have different crystal lattices, the film will grow strained in order to match the lattice structure of the substrate, while a misfit stress is built up in the film. At a specific film thickness, stress will be partially released by the generation of energetically more favourable dislocations. Thus, the system is governed by the force balance between film stress and the tension of dislocations, which can be considered as two competing species. The equation for this specific film thickness, as obtained by Matthews' and Blakeslee's analysis, is given by

$$d_c = \frac{b}{4\pi f_0 \sin\theta\cos\lambda}\left(\frac{1-\nu\cos^2\theta}{1+\nu}\right)\ln\left(\alpha\frac{d_c}{b}\right), \tag{3}$$

with b being the length of the Burgers vector, ν the Poisson ratio, f_0 the lattice misfit, and θ and λ are the angles which define the orientation of the dislocation relative to the lattice. The dislocation core parameter α is usually fitted to experimentally obtained data and may have values typically between 1 and 4.

For further calculation, we introduce the abbreviations

$$A = \frac{b}{4\pi f_0 \sin\theta\cos\lambda}\left(\frac{1-\nu\cos^2\theta}{1+\nu}\right) \tag{4}$$

$$B = \frac{\alpha}{b} \tag{5}$$

and then obtain Eq. (1).

Using the transformation $d_c \to u = Bd_c$ and the abbreviation $-1/AB = k$, we obtain the simple implicit equation

$$ku + \ln u = 0. \tag{6}$$

We begin to solve Eq. (6) using the *Ansatz*

$$u = \frac{1}{k}W(k). \tag{7}$$

Inserting Eq. (7) into Eq. (6) yields

$$W(k) + \ln\left(\frac{1}{k}W(k)\right) = 0. \tag{8}$$

Rearranging Eq. (8) and employing an exponential in Eq. (10)

$$W(k) - \ln k + \ln W(k) = 0, \tag{9}$$

$$\exp(W(k) - \ln k + \ln W(k)) = 1, \tag{10}$$

$$\exp W(k)W(k) = k \tag{11}$$

we find that Eq. (11) represents the definition of the Lambert W-function, as already established in Eq. (2).

Therefore, our *Ansatz* in Eq. (7) is justified.

The exact solution for the critical film thickness is after resubstitution for k

$$d_c = -AW\left(-\frac{1}{AB}\right) \tag{12}$$

and, after resubstitution for A and B:

$$d_c = \frac{-b}{4\pi f_0 \sin\theta\cos\lambda}\frac{1-\nu\cos^2\theta}{1+\nu} \times W\left(\frac{-4\pi f_0 \sin\theta\cos\lambda}{\alpha}\frac{1+\nu}{1-\nu\cos^2\theta}\right). \tag{13}$$

Lambert's W-function is a complex and multi-valued function with an infinite number of branches, only two of them having real values. If x is real, then for $-1/e \leqslant x < 0$ there are two possible real values of $W(x)$, as displayed in Fig. 1.

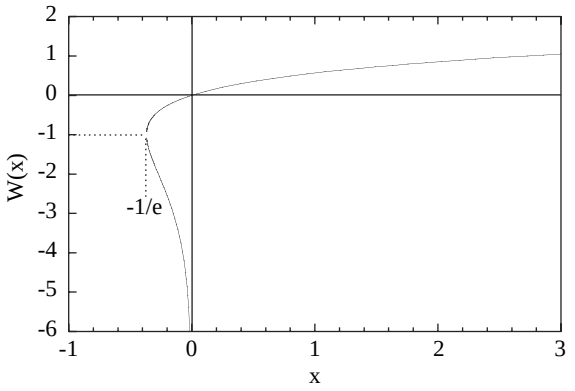

Fig. 1. The real valued Lambert W-function with branches W_0 and W_{-1}.

The branch satisfying $-1 \leqslant W(x)$ is denoted $W_0(x)$ and referred to as the principal branch in the literature; the branch satisfying $W(x) \leqslant -1$ is denoted $W_{-1}(x)$ [3].

We mention that the W-function can be differentiated

$$W'(x) = \frac{W(x)}{x(1+W(x))} = \frac{e^{-W(x)}}{1+W(x)} \tag{14}$$

and also integrated

$$\int W(x)\mathrm{d}x = x\left(W(x) - 1 + \frac{1}{W(x)}\right) = (W(x)^2 - W(x) + 1)e^{W(x)}. \tag{15}$$

For practical applications, as we have in mind here, we have to recall that there are two possible values of $W(x)$ for $-1/e \leqslant x < 0$. To find the branch of $W(x)$ that correctly describes the evolution of the critical thickness additional plausible considerations are required.

The argument of the W-function as well as the prefactor have always the same sign, either negative or positive. Therefore, the critical thickness d_c becomes always positive as expected. We identify $W_{-1}(x)$ (Fig. 1) as the branch that correctly describes the evolution of the critical thickness as a function of the lattice misfit f_0, because a misfit $f_0 = 0$, that is, $x = 0$, must correspond to $d_c = \infty$ (and not $d_c = 0$). An increasing misfit must cause a decreasing critical thickness.

We demonstrate how the W-function is applied to obtain an explicit expression for the critical thickness as a function of the composition of the substrate:

Consider the critical thickness of nickel thin films as a function of the misfit, as it occurs when growing it on a single crystal made from a binary alloy such as $Cu_xAu_{(1-x)}$, its surface being indexed as (0 0 1). Thus, the critical thickness can be expressed as a function of the concentration of copper. The lattice parameter a of $Cu_xAu_{(1-x)}$ is obtained by Vegard's rule [8], while we neglect that for some systems and some compositions this rule might not hold

$$a = a_{\mathrm{Cu}}x + a_{\mathrm{Au}}(1-x) \tag{16}$$

with $a_{\mathrm{Cu}} = 3.615\,\text{Å}$ and $a_{\mathrm{Au}} = 4.078\,\text{Å}$. The lattice misfit f in the copper film is defined as

$$f = \frac{a - a_{\mathrm{Ni}}}{a_{\mathrm{Ni}}}, \tag{17}$$

where a_{Ni} being the lattice parameter of nickel, 3.5238 Å.

Eqs. (16) and (17) can be inserted in Eq. (13) accordingly. For further calculation we take for the Poisson ratio of nickel $\nu = \frac{153}{400}$, for the Burger's vector $a_{\mathrm{Ni}}/2[1\,1\,1]$, for the dislocation core $\alpha = 3$, and we assume 60° dislocations so that $\theta = \pi/2$, $\lambda = 0$. The result is displayed in Fig. 2, which shows the critical thickness of nickel grown on a copper/gold single crystal, as a function of the concentration of copper.

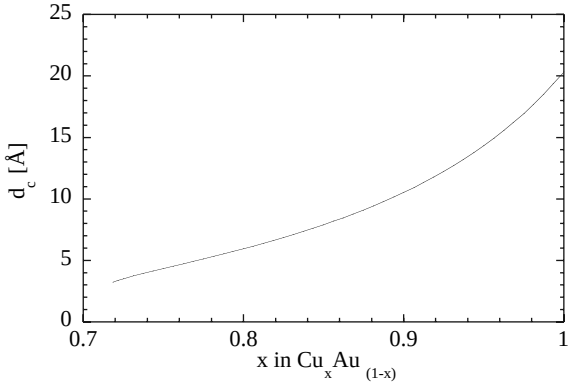

Fig. 2. Critical thickness for the onset of misfit dislocations in nickel as a function of copper content x in the substrate.

The case $x = 1$ represents the pure copper substrate with a lattice constant of 3.615 Å, which allows for the smallest lattice misfit of 2.59% and thus for the largest critical thickness of about 20 Å.

With decreasing x, the gold content in the substrate increases, and so does the lattice parameter, after Vegard's rule. Consequently, the lattice misfit increases to about 15%, and the critical thickness decreases to about 2.8 Å.

3. Conclusions

Eq. (13) represents an expression for the critical film thickness as of Matthews' and Blakeslee's definition in an exact and algebraic closed and explicit form. By reformulating the equation in W form we take advantage of the whole theory of the W-function, which might be useful for understanding of the solution.

Within the framework of the concept of exactly solvable growth models [4–6], the present case can be considered as the competition of two species, that is, the misfit stress in the film versus the tension of the dislocation. The balance of the two forces finds its manifestation in the occurrence of the critical thickness.

Though the new representation of the critical thickness does not provide any new physics of the underlying principles, it enhances the transparency of the system particularly for cases where the evolution of the critical film thicknesses is considered as a function of variables, such as the lattice misfit f.

The new expression for the critical film thicknesses requires knowledge about the Lambert W-function.

One might argue that it is possible to present an explicit solution for any implicit equation, once one has defined an appropriate novel function.

However, in the case of the Lambert W-function, there exists a fast and simple computer algorithm [9] to compute this function to arbitrary accuracy, as for other known transcendental functions like sin, exp, and so on. This algorithm is based on Halley's iteration, which converges faster than Newton's method. It is also more reliable because it takes asymptotic behaviour into account, which guarantees higher accuracy for very small and very large arguments. Also, various mathematical computer programs such as Maple and Mathematical have already implemented this function. Additionally, by reformulating our problem in terms of W, a large body of available theory immediately becomes applicable to this problem.

The closed form of d_c can be easily implemented in other symbolic relations, for instance in magnetoelasticity [7] or in works which employ the epitaxial Bain path [7], but also for modeling work on semiconductor heterostructures.

References

[1] J.E. Matthews, A.E. Blakeslee, J. Crystal Growth 27 (1974) 118.
[2] The Science Citation Index for Ref. [1], as of May 6, 2001.
[3] S.R. Valluri, D.J. Jeffrey, R.M. Corless, Can. J. Phys. 78 (2000) 823.
[4] K.M. Briggs, Some exactly solvable growth problems, in preparation.
[5] A. Braun, M. Bärtsch, B. Schnyder, R. Kötz, Chem. Eng. Sci. 55/22 (2000) 5245.
[6] A. Braun, H.-G. Hermanns, A. Wokaun, Analytical solution to a growth problem with two moving boundaries, J. Phys. A, submitted for publication.
[7] A. Braun, B. Feldmann, M. Wuttig, J. Magn. Magn. Mater. 171 (1997) 16.
[8] L. Vegard, Z. Phys. 5 (1921) 17.
[9] Source code: `www.btexact.com/people/briggsk2/LambertW.c`.